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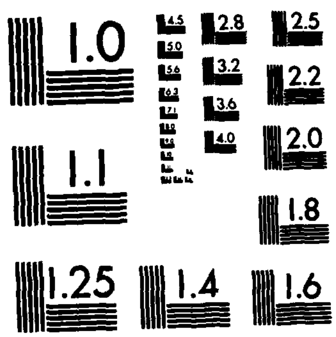

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Nonexponential Decay in Relaxation Phenomena and the Spectral Characteristics of the Heat Bath

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ABSTRACT

After a brief survey of the characteristics of a heat bath and its role in relaxation phenomena leading to the familiar exponential decay, it is argued that the nonexponential form found commonly in many condensed matter systems indicates that the energy spectrum of the heat bath plays a crucial part in these phenomena. In equilibrium statistical mechanics, the mean energy of a heat bath determines the temperature of a system placed in contact with it. We show that the relaxation of a system placed in contact with this heath bath is determined by the distribution of the energy level spacings for level spacings small as compared to the mean spacing. After presenting arguments in favor of a linear behavior of this distribution, we show, in a somewhat heuristic way, that the resulting relaxation function has a nonexponential form.

I. INTRODUCTION

Experimental data on relaxation phenomena in diverse areas of condensed matter physics are quite generally found to exhibit slower than exponential decay for long times in the form (see ref. 1. and references therein)

$$\exp\left[-a(t/\tau_s)^b\right], \quad a>o, \; o<b<1 \tag{1}$$

where τ_s is a characteristic time in the system. Traditionally[2,3] the residual part of relaxing quantities are discussed either in terms of a pure exponential decay

$$\exp[-t/\tau] \tag{2}$$

where τ is the "relaxation time", or in terms of a superposition of such terms with a distribution of τ. $\tau_s \neq \tau$ in general but there is a relation between them (see Ref. 1.). (References 4,5,6 are a representative set selected at random out of a large number of papers on this subject. See Ref. 1. for more citations). In this framework, τ is obtained from a calculation of the time-independent transition rate given by the Golden Rule. The physical picture of relaxation here is that the system which is relaxing and which is described by a Hamiltonian, H_S, is in contact with a heat bath which is a much larger system described by a Hamiltonian, H_B, and which is not affected by the interaction (H_{BS}) with the relaxing system. This interaction is supposed to be "weak" and leads to relaxation. The equilibrium properties of the given system in contact with the heat bath are determined by the temperature of the bath, which in turn is just the mean energy associated with the bath.[7,8]

An alternative scheme to derive (2) is the master equation approach (see for example Ref. 8). Here again the time-independent transition rate is employed in setting up the master eqation. The physical model for relaxation is however still the same - system, heat bath, and their mutual (weak) interaction. An elementary discussion of the exponential decay in stochastic processes and in quantum mechanics may be found in Merzbacher's book,[9] and an extensive discussion of the role of time independent transition rates is found in Tolman[7].

Now, since exponential decay is not observed, and the decay law given by (1) is more a rule than an exception, it is natural to seek an explanation for this behavior by examining in more detail the origin of the time-independent transition rate. This shift of emphasis from time independent transition rate (TITR) to time dependent transition rate (TDTR) in order to arrive at (1) has been emphasized recently by Teitler et.al.[10] from phenomenological considerations of rate equations, and from general considerations based on the Paley-Wiener theorem by Ngai et.al.[11] It was also recognized early in the development of the TDTR that using the Golden Rule[1] required a linear dependence of the level spacing density for low spacings in order to obtain the long time behavior of the form given by Equ. (1). The purpose of the present paper is to suggest that the heat bath be described by any chaotic quantum system. The reasons for this suggestion are given in Sec. II. In Sec. III, we set up the calculational scheme for computing TDTR and obtain the required time dependence in terms of the slope and cut off of the linear law for the level spacing distribution at small spacings of the heat bath. In Sec. IV a brief summary of the results obtained is given.

II. SPECTRAL CHARACTERISTICS OF THE HEAT BATH

We are primarily interested in the long time relaxation of any physical property of a system, for example a dielectric or a mechanical property. What is involved in a relaxation process is a readjustment of the system in such a way that there is no transport phenomenon accompanying it.[12] In general terms, the system that is undergoing relaxation is described by a Hamiltonian H_S, and it is supposed to be in contact with a much larger system described by a bath (or reservoir) hamiltonian H_B. The interaction between the two, whose hamiltonian is H_{BS}, is assumed to be weak and is supposed to induce the process of relaxation in the physical quantity of interest in the system. It is important to realize that the bath system is large compared to the system that is under investigation so that while the bath is not affected by the interaction with the system, its effect on the system is paramount.

The precise nature of the heat bath is left unspecified except for stating that the system in contact with it acquires its temperature in the equilibrium situation. In the conventional approach, the details of the bath hamiltonian are not important even though it is recognized to have an almost continuous energy spectrum, by virtue of its enormous size.[7] Also, in the final analysis, the bath variables do not occur in the description of the system so that one averages over these variables in computing properties of the system. Since only the temperature of the bath enters the picture the only relevant entity appearing in this picture is the mean energy of the bath system, which is kept fixed, thus determining the temperature. It is clear from this description of the heat bath that a detailed knowledge of H_B is not required, except for its temperature and the obvious observation that H_B has an almost continuous spectrum bounded from below with a finite mean. One of the approaches to the theory of relaxation is to construct the density matrix associated with the system plus bath in the presence of H_{BS} and integrate out the bath variables by tracing over all the states of the bath, leaving behind a residual density matrix for the system, which can be used to compute any property, P(t), of the system, by calculating the appropriate average of the operator representing P(t) over the residual density matrix. Feynman and Vernon[13] have given a formal path integral representation for this density matrix and Fano[14,15] has given an expression in the interaction representation.

It may not be out of place here to point out that there are circumstances when the nature of the heat bath is known purely from the physical consideration of the energy or time domain one is examining. For certain electronic properties, phonons (the motion of the crystal lattice) are the relevant heat bath system and so for excitations involving frequencies of the order of 10^{13} Hz and above, the phonon excitations determine the relaxation properties upto times of order 10^{-13} sec and here the relaxation rate is essentially exponential. When one waits for longer times, say 10^{-10} sec, the phonon excitations are not relevant any longer and the relevant bath system must be something else. It must be pointed out however, that the phonon system is itself imbedded in the new bath system which we are proposing so that there is a common temperature for all the entities making up the system. Thus we may picture a hierarchy of heat baths, each imbedded in the other, so that they all have a common temperature and each is a relevant heat bath in the appropriate time regime. This nesting of heat baths leads to different time dependences of the relaxing entity in different time domains. What we are interested in for the present work is the relatively long time domain such as 10^{-10} sec and lower, where the usual known excitations become irrelevant and a new mechanism is called for. In the present paper we are concerned only with this regime. It must be stressed that the theoretical formalism given by Fano[14,15] is applicable quite generally to all these situations.

In the traditional description[7] no mention of the nature of the heat bath energy spectrum is made except for its being continuous. Thus we describe the system undergoing relaxation by means of the Hamiltonian

$$H = H_S + H_B + H_{BS} \tag{3}$$

where H_S, H_B, and H_{BS} are the hamiltonian for the system, bath, and their mutual interaction. One may then compute the density matrix of the entire system, given that at time t=0 the system and the bath are not interacting, and have been prepared such that the system is in some preassigned state and the bath is in thermal equilibrium.

As a model for the heath bath we shall adopt any large quantum system whose classical motion is irregular. For such systems Berry has shown in a series of elegant papers[16-20] that the quantum levels are fairly regularly distributed and that the probability density P(S) for the spacings between neighboring levels has the asymptotic form

$$P(S) \approx \alpha S \qquad\qquad (0<S<<\delta) \tag{4}$$

provided S is small as compared to the average spacing δ . For larger values of S the spacings distribution P(S) goes through a maximum to decay to zero at large values of S. These details depend on the precise choice of the quantum system, but the linear behavior of the spacings distribution appears to be universal, i.e. it holds for a "generic" chaotic quantum system.

It is remarkable that the same linear behavior (4) of the spacings distribution is found if the heat bath is described by means of a random matrix hamiltonian. The hamiltonian for the heat bath is very complex so that we may replace it by a statistical description. For the determination of equilibrium properties of a system in contact with the heat bath, only the average of the bath hamiltonian is needed. Hence one can try to use a "Gaussian Orthogonal Ensemble" (GOE) for the random matrix for describing the heat bath because we take the bath system as being time reversal invariant. Since only the mean value of the bath Hamiltonian is required for equilibrium properties, we use a "canonical ensemble" in setting up its density matrix. We now observe that the GOE has known average spectral properties, which we employ in our analysis of the Golden Rule in determining the TDTR induced by the bath in the system. For a description of the philosophy and the theory of random matrix hamiltonians one may refer to Porter's collection of papers and his clear introductory summary[21], Mehta's book[22], and a more recent review by Brody et. al.[23].

It is remarkable that both models for the heat bath which we have considered in this section, (a) an irregular quantum system and (b) a random matrix hamiltonian, lead to the same linear behavior of the spacings distribution. We feel that (a) is the physically correct model for a heat bath and that the random matrix is just a convenient way to simulate an irregular quantum system. It may not be out of place here to conjecture that the random matrix hamiltonian may indeed be a very accurate model for a generic irregular quantum system if one coarse grains the energy spectrum. A hint of this equivalence may be found when one compares the average density of states for the quantum version of Sinai's billiard [19] with the middle part of the semi-circular law appropriate for the density of states of a random matrix.[21-23]

III. CALCULATION OF THE TIME DEPENDENT TRANSITION RATE

The calculation proceeds in three steps: (i) set up a rate equation for the physical quantity that is undergoing relaxation in terms of a TDTR; (ii) compute the TDTR; and (iii) solve the rate equation once the TDTR is determined. Our goal is to examine the long time limit of the time dependence and so there is much simplification that can be made right from the start. A formal justification of step (i) is given elsewhere[24]. We are concerned here mainly with step (ii) and calculate the transition rate using the Golden Rule with proper attention paid to the types of interactions that could be suggested for H_{BS}. Having done this, we then invoke the cumulant expansion technique[14] or equivalently the linked cluster scheme to calculate the TDTR essentially to all orders in H_{BS}. This is in the same spirit as in the binary correlation approximation.[23]

We are thus led to consider the survival probability of a state of the system when it interacts with the bath. Let the heat bath have states $|b\rangle$ and the relaxing system two representative states $|1\rangle$ and $|2\rangle$. At t=o, the combined system (S + B) is in a state $|s,b\rangle$ where $|b\rangle$ is some state with its energy in the range $E - \Delta < E_b < E + \Delta$. For a "good" heat bath, in general, one does not know the state $|b\rangle$ apart from the fact that it has an energy E_b in some energy region $(E-\Delta, E+\Delta)$. The standard <u>Golden Rule result</u>[9] for the probability that the total system is in the state $|s'b'\rangle$ at time t given the initial state is $|sb\rangle$ is

$$|C_{s'b'}(t)|^2 = \frac{2}{\hbar^2} |V_{sb,s'b'}|^2 (1-\cos \omega_{sb,s'b'}t)\, \omega^{-2}_{sb,s'b'} \tag{4}$$

where $\hbar\, \omega_{sb,s'b'} = E_{s'} + E_{b'} - E_s - E_b$, with E_s, the system energy. The matrix element of the system-bath coupling Hamiltonian H_{BS} between the states $|sb\rangle$ and $|s'b'\rangle$ is denoted by $V_{sb,s'b'}$.

In order to obtain the total transition probability $Q_{s,s'}(t)$ for the system to go from state $|s\rangle$ to state $|s'\rangle$ irrespective of the state of the heat bath we sum over $|b'\rangle$ and average over $|b\rangle$. This gives for the probability to find the system in state $|s'\rangle$ at time t

$$Q_{s,s'}(t) = \frac{2}{\hbar^2 B} \sum_{b,b'} |V_{sb,\, s'b'}|^2 (1-\cos \omega_{sb,s'b'}t)\, \omega^{-2}_{sb,s'b'}, \tag{5}$$

where B denotes the number of heat bath states in $(E-\Delta, E+\Delta)$. In order to express the relaxation function $Q_{s,s'}(t)$ in terms of the spectral characteristics of the heat bath one now proceeds, in a somewhat heuristic fashion, as follows.

Firstly, as the system-bath coupling will depend only on the global properties of the heat bath the matrix element $V_{sb,s'b'}$, will be practically independent of the choice of $|b\rangle$ and $|b'\rangle$ in the energy range $(E-\Delta, E+\Delta)$. This enables us to put

$$V_{sb,s'b'} \simeq V_{s,s'} \tag{6}$$

and to bring the constant $V_{s,s'}$ outside the summation sign in (5).

Secondly, we note that the resulting sum over b' will be independent of the choice of b. Hence the average over b is trivial and (5) can be written as the sum

$$Q_{s,s'}(t) = \frac{2}{\hbar^2} |V_{s,s'}|^2 \sum_{b'} (1-\cos\omega_{sb,s'b'}t)\ \omega^{-2}_{sb,s'b'}. \tag{7}$$

In a relaxation process, the transition occurs from a state of the system to another state of the system which is essentially degenerate with it and so $\hbar\omega_{sb,s'b'} = E_{b'} - E_b$. The sum (7) can now be written as an integral

$$Q_{s,s'}(t) = 4\ |V_{s,s'}|^2 \int_0^\infty (1-\cos \varepsilon t/\hbar)\ \varepsilon^{-2}\ \rho(\varepsilon)\ d\varepsilon, \tag{8}$$

where $\rho(\varepsilon)\ d\varepsilon$ denotes the average number of heat-bath quantum states with energies in $(E_b+\varepsilon,\ E_b+\varepsilon+d\varepsilon)$ given a heat-bath level at E_b. For ε large as compared to the average level spacing δ one has $\rho(\varepsilon) \simeq \delta^{-1}$. On the other hand, for $\varepsilon \lesssim \delta$ we can use the result (4) for the distribution of level spacings of an irregular quantum system and put $\rho(\varepsilon) \simeq \alpha\ \varepsilon$.

Thirdly, we use the following qualitative form for ρ suggested by the preceeding remarks

$$\rho(\varepsilon) = \alpha\ \varepsilon \qquad 0<\varepsilon<\varepsilon_0, \tag{9a}$$

$$\rho\ (\varepsilon) = \delta^{-1} \qquad \varepsilon_0 < \varepsilon < \infty, \tag{9b}$$

$$\varepsilon_0 = (\alpha\delta)^{-1}. \tag{9c}$$

The calculation of the integral (8) is now straightforward, and leads to the asymptotic behavior

$$Q_{s,s'}(t) \simeq 4\ \alpha |V_{s,s'}|^2\ \ln(\tfrac{t}{\tau}) + \ldots\ldots, \qquad (t \gg \alpha\delta\hbar), \tag{10a}$$

$$\tau = \alpha\ \delta\ \hbar\ \exp(-1-\gamma). \tag{10b}$$

The dots in (10a) denote terms that vanish for $t \to \infty$, and the constant γ=0.5772 is Euler's constant. Of course, for small values of t the quantity $Q_{s,s'}(t)$ will be proportional to t^2.

The calculation outlined above amounts to lowest order perturbation theory. It was shown by Fano[14] that when one proceeds in a rigorous fashion one obtains a cumulant expansion for the transition probability, which essentially leads to an exponentiation of the Golden Rule result (10)

$$Q_{s,s'}(t) \simeq (t/\tau)^b, \qquad (t \gg \tau), \tag{11a}$$

$$b = 4\,\alpha |V_{s,s'}|^2. \tag{11b}$$

With a time dependent transition rate of this form the solution of a rate equation will lead to terms of the form (1), with 0<b<1, for otherwise, with b>1, one has faster than exponential decay.

IV. CONCLUDING REMARKS

We have shown, in a somewhat heuristic way, how the fine-grained spectral characteristics of the heath bath determine the form of the relaxation function. It is remarkable that the only quantities which enter are the average spacing δ, the average system-bath matrix element $V_{s,s'}$, and the slope α of the spacing distribution at small spacings. The linear behavior of this distribution at small spacings is a generic feature of irregular quantum systems[16-20], hence an irregular (chaotic) quantum system is a universal model for a heath bath.

Our considerations also show that one can use the Gaussian orthogonal ensemble of random matrices to simulate a chaotic quantum system, and hence to model a heath bath. This might also explain the success of random matrix theory in nuclear physics and other branches of physics.

The main motivation of the work reported in this paper was to make the concept of the heat bath, which up till now inhabited the literature on statistical physics as an almost featureless entity, more specific and to determine which of its properties enter into the physics of relaxation.

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